

EVALUATION OF CONTAMINATED AIR VELOCITY ON THE FORMALDEHYDE REMOVAL EFFICIENCY BY USING A BIOTRICKLING FILTER REACTOR

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ABSTRACT:

Introduction: Formaldehyde is a toxic, carcinogen, mutagen and teratogen compound that is widely released into the atmosphere worldwide. The toxicity effect of formaldehyde on microorganisms is a serious obstacle on the path of using biological treatment methods. The main objective of this study is to evaluate the effect of air velocity on the efficiency of a Biotrickling Filter Reactor (BTFR) for removal of formaldehyde from an air stream.

Materials and methods: A BTFR and Bioscience Laboratory Respirometer were employed for continuous and batch experiments, respectively. Three gas flow rates including 90, 291 and 1512 L/h were used to find out the effect of velocity on the formaldehyde removal efficiency of the BTFR. Monod model was modified to be capable of predicting the BTFR cases having very high formaldehyde removal efficiency.

Results: The results showed that for the gas flow rates of 90, 291 and 1512 L/h in BTFR, formaldehyde removal efficiency of 95, 97 and 99% were achieved, respectively. These results showed that higher air flow velocity lead to higher removal of formaldehyde from air in a BTFR. A very slow formaldehyde removal was observed during batch experiment where the gas velocity is set equal zero.

Conclusions: This study demonstrated that the mass transfer in gas phase is an important step in formaldehyde treatment in a BTFR. Very slow formaldehyde removal in the batch system which can be ascribed to the very low mass transfer rate in gas phase confirms the fact that this rate is a controlling step in overall removal rate in BTFR.

INTRODUCTION

Continuous discharge of chemicals through wastewater, flue gas or solid waste has produced such a serious impact on the environment that

has hindered sustainable development of industries [1]. A large number of research cases aiming at development of new methods or improvement of the existing ones for the management

and/or treatment of industrial wastes have been published so far. Formaldehyde appears in a diversity of gaseous industrial waste being released through ventilation air streams and/or stack gases [2]. Formaldehyde is known as toxic and mutagenic compound which is considered as a cause of fetal birth defects [3]. Formaldehyde is highly soluble in water with the solubility of about 40 g/L [4]. Although formaldehyde has harmful effects on microorganisms, many researchers have tried to use biological methods for removing formaldehyde from the air [1, 3-7]. Biological methods have been proved to be able to remove formaldehyde using both aerobic and anaerobic methods [8]. Various mechanisms have been proposed to explain the anaerobic decomposition of formaldehyde. According to previous studies, formaldehyde is biologically converted to formic acid and methanol. Then, formic acid and methanol is converted to methane and carbon dioxide by microorganisms [9]. Another mechanism has been suggested by researchers that showed formaldehyde is transformed to volatile fatty acids, especially formic acid being converted to acetic acid. Finally, acetic acid is converted to methane and carbon dioxide through anaerobic bioreactions [10]. The aerobic removal of formaldehyde occurs in two ways. The first mechanism involves the conversion of formaldehyde into methanol and formic acid by formaldehyde dismutase enzyme ($2\text{HCHOCH}_3\text{OH} + \text{HCOOH}$) released by microorganisms [2]. In the second mechanism, the conversion of formaldehyde to formic acid takes places by another enzyme named as dehydrogenase (HCHOHCOOH) [7]. In this case, methanol and formic acid are converted into water and carbon monoxide through biological activities.

Five different biological scan system serve as an aerobic reactor to remove gaseous pollutants. These systems include 1) biofilters (BF), 2) biotrickling filters reactors (BTFR), 3) bioscrubbers (BS), 4) biomembrane reactors (BMRs), and (5) suspended growth reactors (SGRs) [11]. BTFRs have been gained attention due to giving several advantages, i.e. the ability of acid gas

treatment, having lasting supporting materials and producing low pressure drop [3]. BTFR is a suitable method for removal of chemicals with high solubility in water such as volatile organic compounds (VOCs). Traditionally, biofilters are used to treat exhausted gases with VOC concentrations below 10 mg/L [6]. BTFRs are usually packed with synthetic supporting materials. Microorganisms can grow as biofilm layer on the surface of supporting materials. Microorganisms require energy, carbon, nutrients and water resources for their survival [12]. In a BTFR reactor contaminated water contains carbon sources. A nutrient solution which is composed of water and several necessary chemicals is circulated through the packed bed to supply biofilms with these chemicals and water [2]. Previous studies showed that wastewater containing soluble can be treated by the means of two mechanisms in BTFRs [7]. The most of the air pollutants are dissolved into nutrient solution and are removed by the activities of microorganisms in the biofilm. Another part of pollutants can be removed directly by some microorganisms such as fungi [7]. Although several studies were carried out to investigate the removal of VOCs from contaminated gases using BTFR system, there is no study to compare the batch and continuous operation of this system. Also, no reliable equation has been already presented in the literature for prediction of VOCs removal by a BTFR. The present study is concerned with the evaluation of BTFR performance in batch and continuous modes to remove formaldehyde from an air stream. This evaluation ends up with development of a mathematical correlation between formaldehyde removal efficiency and gas retention time.

MATERIALS AND METHODS

Continuous system

The present study was conducted with a laboratory-scale BTFR (see Fig.1). The small pieces of polyurethane tube with 1 cm long and 0.5 cm in diameter were used as a support material. The small pieces of polyurethane tube with 1 cm long

and 0.5 cm in diameter was used as a support material. This type of support provides BTFR with 90% void percentage in the bed. As it can be seen in Fig.1, BTFR has four sampling ports 1, 2, 3 and 4 located at 5, 10, 10 and 41 cm from the bottom of the biofilter beds, respectively. Fig.2 shows a schematic picture of the employed BTFR. Aerobic sludge (AGS) collected from municipal wastewater treatment plant in Isfahan, Iran, was used to inoculate BTFR in the startup phase. For our tests, the nutrient solution flow rate is adjusted at 50 L/h using a pump equipped with a switch with a manipulatable mechanical clock. Some elements such as carbon, nitrogen, phosphorus, magnesium, potassium, calcium, iron, chloride and manganese are essential for microbial growth. The nutrient solution containing a sufficient quantity of minerals with a given for-

mulation (Table 1) supply required moisture of during the experiments. It has been reported that the optimum pH value of nutrient solution for removal of formaldehyde in BTFR is around 7 [12]. Thus, the pH of nutrient solution was adjusted on 7.

Table 1. Nutrients solution formulation

Name of materials	Amount (g/L)
MgSO ₄	0.1
KH ₂ PO ₄	0.5
CaCl ₂ .2H ₂ O	0.01
FeSO ₄	0.001
NH ₄ Cl	1
K ₂ HPO ₄	0.5
MnSO ₄	0.001

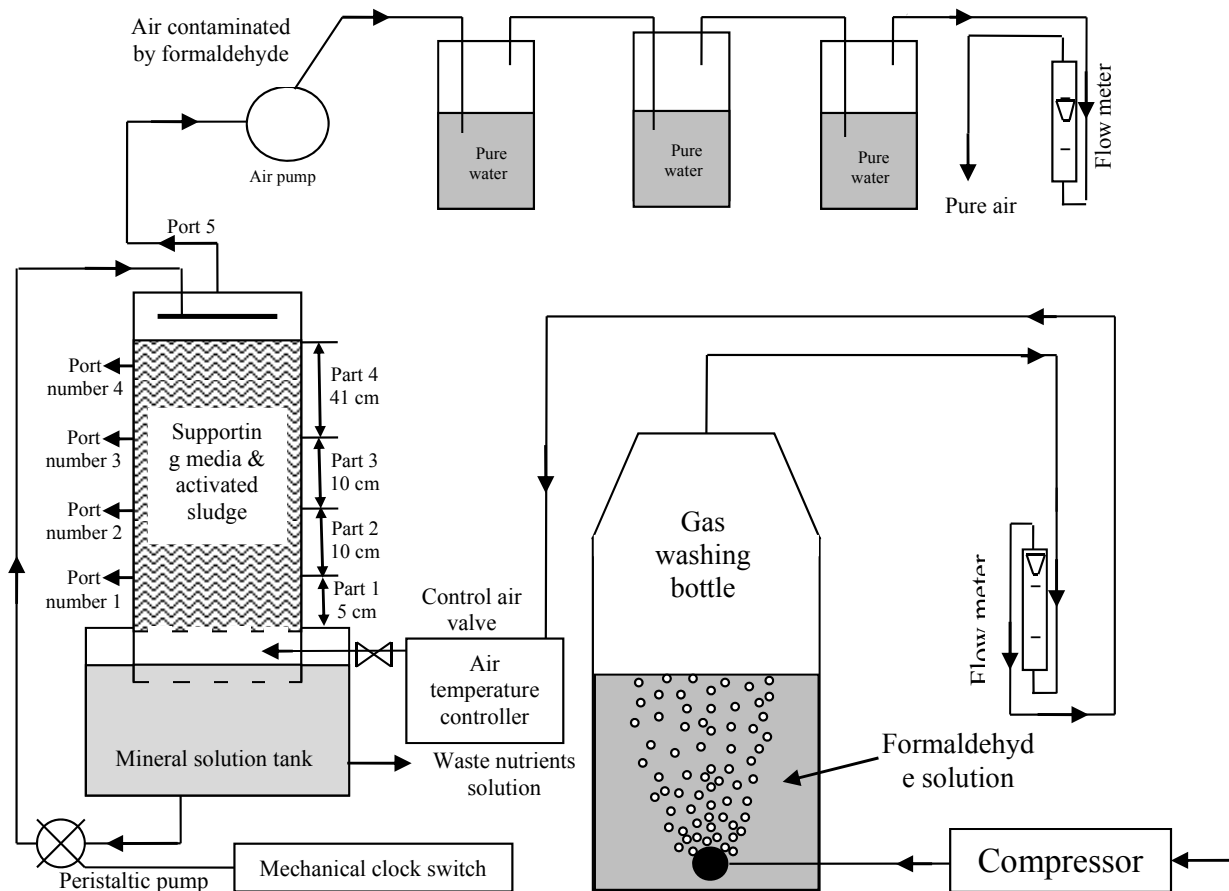


Fig.1. Schematic of an artificial BTFR and contaminated air flow (SCAS) continuous production systems

In the first phase, an attempt was made to adapt microorganisms to formaldehyde by adding formaldehyde to the nutrient solution as a carbon source for 80 days. Formaldehyde concentration in nutrient solution was determined using chemical oxygen demand (COD) measurement. In order to evaluate the effect of gas velocity on BTFR performance, the formaldehyde removal efficiencies were determined for the volumetric air flow rates (VAFR) of 90, 291 and 1512 L/h. The formaldehyde concentration in input SCAS remained constant at 490 mg/L during all experiments. In order to determine the formaldehyde concentration distribution through gas phase, air samples were taken from the ports 1 to 4 using a vacuum pump, and they passed through a back wash system for a given period of time (see Fig.1). The COD of the remaining solutions in the back wash bottles were measured and it was correlated to the gas-phase formaldehyde concentration using a calibration curve. The temperatures of gas stream at inlet, middle and outlet of biofilter bed were measured steadily using a digital thermometer. All of the above-mentioned experiments were replicated twice for precision assurance.

Batch system

A bioscience laboratory respirometer model DI-2000 was used to study formaldehyde removal in a batch reactor system. Fig.2 shows the schematic of bioscience laboratory respirometer. This system is able to produce oxygen to prevent an anaerobic condition. In this work, 30 pieces of supporting materials covered with the biofilm containing the predominant microorganisms were placed into an airtight bottle of 1000 mL. A volume of 1 mL pure formaldehyde was added to this bottle as the sole carbon source, and then the bottle was closed and sealed. Because formaldehyde boiling point is very low (-21°C), it can be evaporated rapidly to increase formaldehyde gas-phase concentration to the desirable level at the beginning of the experiment. Microorganisms consume the formaldehyde absorbed from the gas phase. In the first step of the experiment, an airtight bottle was filled with formaldehyde-free

air. During the experimental run, microorganisms in the bottle produce carbon dioxide and water while consume oxygen and formaldehyde present in the bottle. In order to ensure the activity of microbial metabolisms, it is necessary to supply oxygen into the bottle by an oxygen generator. It is also essential to avoid the pressure drop in the bottle as a result of absorbing CO₂ produced by microorganism activities into the KOH solution, as shown in Fig.2. The amount of oxygen consumption was measured online by bioscience laboratory respirometer .

Developing a model for the BTFR

The rate of formaldehyde removal from a SCAS by the BTFR process was modeled by the following equation [12]:

$$r = r_{max} \frac{C_g}{K_m + C_g} \quad (1)$$

Where r is the rate of formaldehyde removal (mg/L.s), r_{max} is a constant of the maximum rate of formaldehyde removal (mg/L.s), K_m is the saturation constant (mg/L) and C_g (mg/L) is logarithmic average of inlet and outlet concentration, and it is defined as follows:

$$C_g = \frac{C_{in} - C_{out}}{\ln\left(\frac{C_{in}}{C_{out}}\right)} \quad (2)$$

Where C_{in} is the inlet formaldehyde concentration (mg/L) and C_{out} is the outlet formaldehyde concentration (mg/L). A plot of r versus C_g is displayed in Fig.3. Writing formaldehyde mass balance over the whole BTFR as an open system yields the following equation [13]:

$$r = \frac{(C_{in} - C_{out})Q}{V} \text{ or } r = \frac{(C_{in} - C_{out})}{\theta} \quad (3)$$

Where Q is the gas flow rate (L/s); V is the volume of the biofilter (L) and θ is the gas retention time (s).

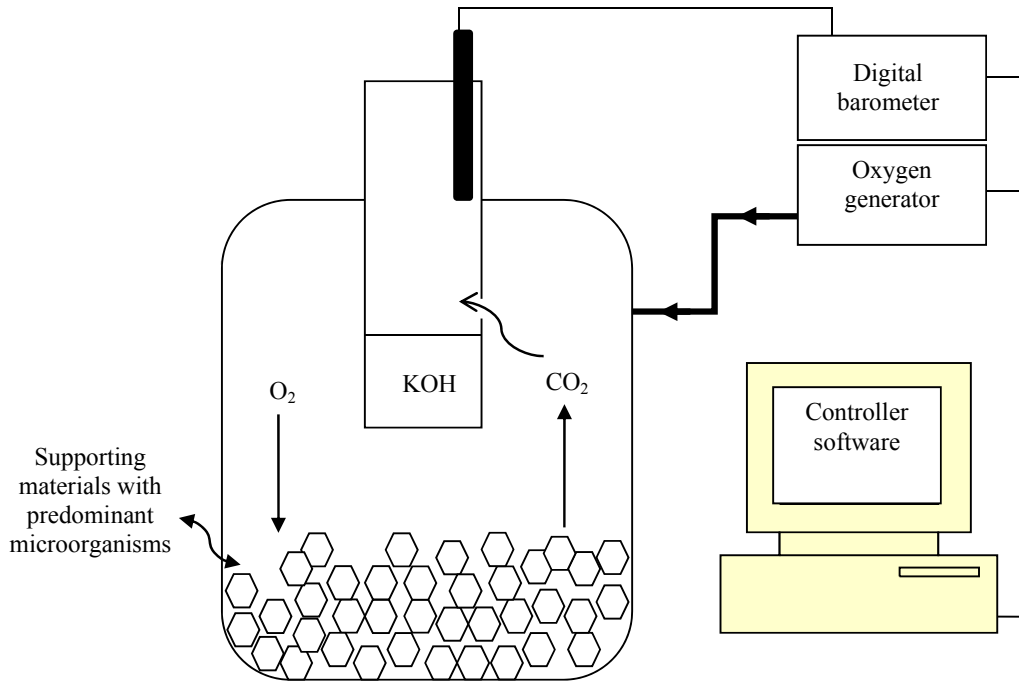


Fig.2. Schematic of bioscience laboratory respirometer

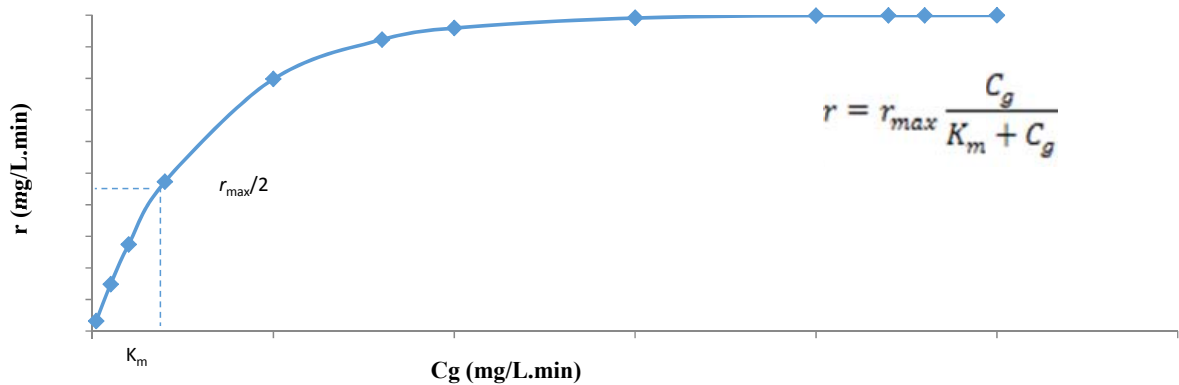


Fig.3. Theoretical model of BTFR treatment process to remove formaldehyde from SCAS

Combining Eqs.(1) and (3) yields Eq.(4) that is:

$$\frac{(C_{in} - C_{out})}{\theta} = r_{max} \frac{C_g}{K_m + C_g} \tag{4}$$

where θ is the gas retention time (s). Rearranging Eq.(4) results in the following equation:

$$\frac{\theta}{(C_{in} - C_{out})} = \frac{K_m}{r_{max}} \times \frac{1}{C_g} + \frac{1}{r_{max}} \tag{5}$$

Because the above equation was not successful to fit the experimental adequately well, it was modified to obtain Eq.(6):

$$\frac{\theta}{(C_{in} - C_{out})} = \frac{K_m}{r_{max}} \times \ln\left(\frac{1}{C_g}\right) + \frac{1}{r_{max}} \tag{6}$$

It is clear that Eq.(6) will show a linear equation, if $(\theta / (C_{in} - C_{out}))$ is correlated with $\ln(1/C_g)$, Y is $(\theta / (C_{in} - C_{out}))$. The slope and interception of such a line are $a (= K_m / r_{max})$ and $b (= 1 / r_{max})$, respectively. Eq.(6) allows us to calculate K_m if the slope

and interception (a, b) of this line were determined through a linear regression analysis. Since $C_{in} - C_{out}$ is defined as $E \times C_{in}$ where E is the efficiency of the BTFR to remove formaldehyde from SCAS [14], rearranging Eq.(6) leads to Eq.(7) which can be used to determine gas retention time of the biofilter for various inlet formaldehyde concentrations of the SCAS.

$$\theta = \frac{(E \times C_{in}) \times \left(\left(K_m \times \ln \left(\frac{1}{C_g} \right) \right) + 1 \right)}{r_{max}} \quad (7)$$

Analytical methods

The contaminated air was passed through three successive bottles containing pure water for a given period of time according to the environmental Protection Agency (EPA) [15] (Figs. 3 and 4). This gas flow was driven using a vacuum pump air pump model champion AAP. Because of high solubility of formaldehyde in water, nearly all formaldehyde content of the contaminated gas was absorbed in water existed in these three bottles. After that time, all three solutions were mixed together and the formaldehyde concentration was measured using COD which was determined using the standard method presented in the literature [16]. Formaldehyde concentration can be correlated to COD according to the reaction $\text{CH}_2\text{O} + \text{O}_2 \rightarrow \text{CO}_2 + \text{H}_2\text{O}$ [4]. Because COD is the oxygen consumed through this reaction, the equivalent formaldehyde concentration can be determined using a mass balance. Also, a hydrocarbon meter was used to verify the accuracy of the above-mentioned method for formaldehyde measurement. As the only hydrocarbon in the synthetic contaminated air is formaldehyde, hydrocarbon meter shows the concentration of formaldehyde directly. The hydrocarbon meter measurement was within the range of 1 to 10000 mg/L. In order to evaluate the biomass concentration in the BTFR 10 pieces of supporting material containing biomass was taken out, and they were placed into a dryer operating at 104 °C for 24 h. Then, the supporting material was washed

by normal HCl solution for 2 h to remove all bio-film. Later on, the cleaned supporting material was placed into a dryer operating at 104 °C for 24 h [17]. The difference between mass of biomass-contained supporting material and the cleaned one can be ascribed to the mass of biomass.

RESULTS AND DISCUSSION

Continues treatment of formaldehyde vapor

Biomass concentrations of BTFR during operation under air flow rates 90, 291, 1512 L/h are 210, 40 and 30 mg/L respectively. Fig.4 shows the variation of formaldehyde removal efficiency of the BTFR system versus the retention time for air volume flow rates of 90, 291 and 1512 L/h. The analyses of the air samples taken from ports 1 to 4 showed that the removal efficiency is 51 to 66 to 76 to 87% for the retention time of 10, 30, 50 and 80 s, respectively. This figure demonstrates that increasing gas flow rate raises the removal efficiency. This can be attributed to an increase in gas velocity which is responsible for enhancing mass transfer rate. This fact reveals that the mass transfer in gas phase is an important step in formaldehyde treatment in a BTFR.

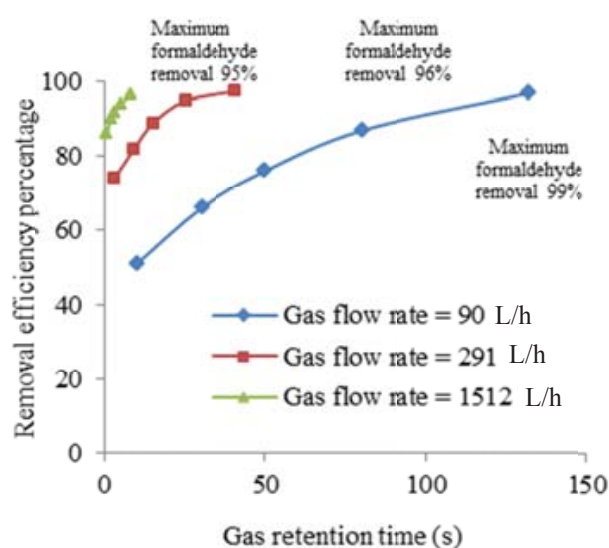


Fig.4. Curves of formaldehyde removal versus gas retention time for three experiments with a VAFR of 90, 291 and 1512 L/h [18]

Batch treatment of formaldehyde vapor

A batch experiment was performed using a bio-science laboratory respirometer model DI-2000 to study the BTFR behavior. The production of CO₂ and the consumption of O₂ by microorganisms during a metabolism process may be estimated using a laboratory-scale respirometric system in a batch reactor, calculated by the following formula: where y is the inverse of oxygen uptake (in l/mg), θ is the gas retention time (in h), α is the empirical constant (in l/mg) and β is the bacterial metabolism coefficient (in L h/mg). The values of α and β as high as 0.717 l/mg and 0.375 L h/mg, respectively, can be verified by plotting y versus $1/\theta$. The correlation between y and $1/\theta$ is very good (, meaning that the parameters α and β have the physical interpretation in the calculation of O₂ needed for bacterial metabolisms.

Fig.6 shows that the oxygen uptake rate is very

small at the beginning of the experiment. The rate of formaldehyde consumed by bacteria increases as time goes by. The consumption (Fig. 5) of O₂ increases suddenly until it reaches at a quite constant value of 1.3 mg/L after 2 h. Based on the following reaction: $CH_2O + O_2 \rightarrow CO_2 + H_2O$, the amount of formaldehyde degraded by bacteria is calculated to be approximately 1.2 mg while the total amount of formaldehyde introduced to the respirometer was 1 mg. This 0.2 mg extra O₂ consumption can be justified by the fact that bacteria have utilized their protoplasm as the carbon source needed for their growth and survival since FA was depleted. The dried-weight amount of biomass was determined both at the beginning and final stages of the experiment as high as 373 and 324 mg, respectively. This 48 mg reduction in biomass verifies the protoplasm utilization hypothesis.

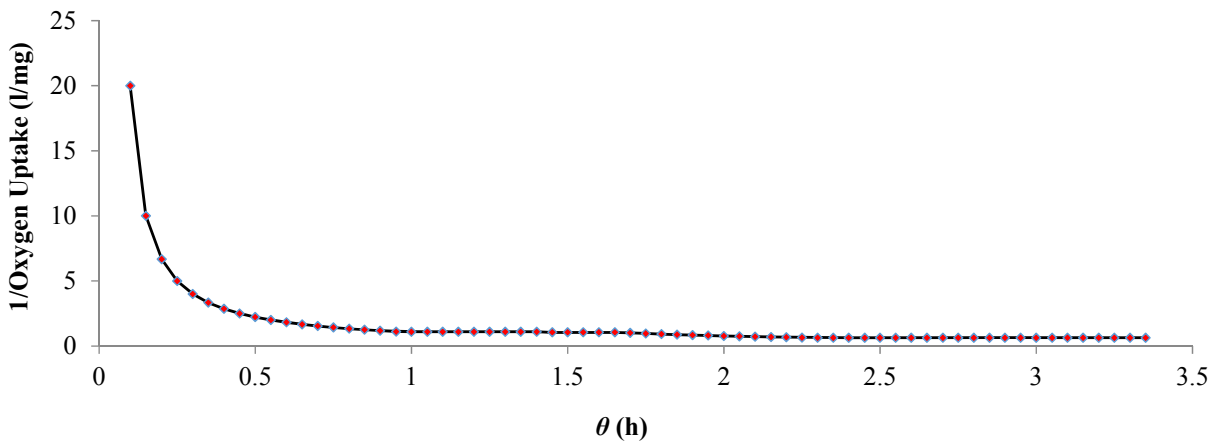


Fig.5. Oxygen uptake in a batch reactor

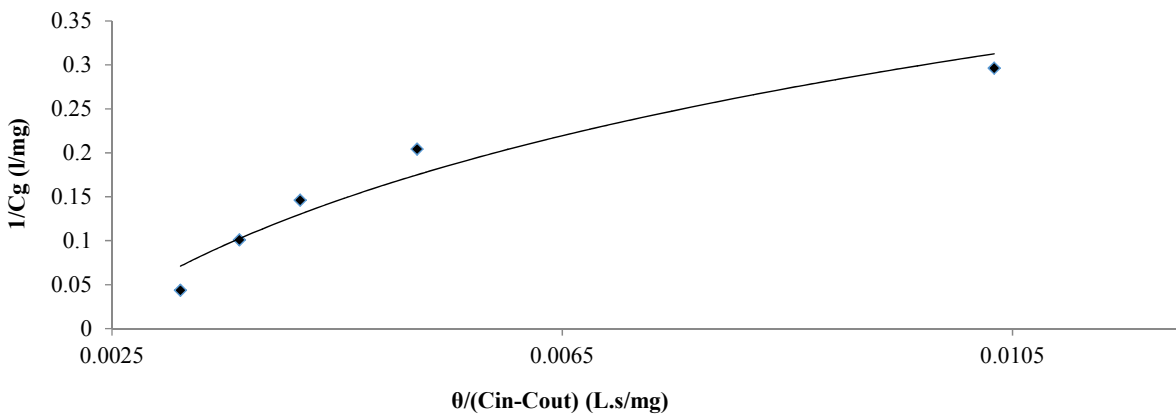


Fig.6. A plot of $\theta / (C_{in} - C_{out})$ versus $1/C_g$; determined by a logarithmic regression

The first data point which was measured after 6 min (360 s) shows the removal efficiency of 4.6%, while for a continuous system the high removal efficiency, very close to unity, would be obtained at such a residence time. This result which can be ascribed to the very low mass transfer rate in gas phase reconfirms the fact that this rate is a controlling step in overall removal rate in BTFR.

BTFR modeling

The theoretical models introduced through Eqs. (5) and (6) provides a better insight into the calculations of gas retention time of the BTFR treatment system, especially for highly purifying BTFR. Regression method was used to find the unknown parameters existed in these models [4, 19].

Eq: $\theta / (C_{in} - C_{out}) = a \times \ln(1/C_g) + b$, with $a = 0.201$ s, $b = 1.2317$ L.s/mg and $R^2 = 0.9433$

A plot (Fig.6) of $(\theta/C_{in} - C_{out})$ versus $1/C_g$ in logarithmic mode determines the values of a and b equal to 0.201 s and 1.2317 L.s/mg, respectively. The correlation coefficient obtained through this fitting was $R^2 = 0.9433$ demonstrating a good agreement of the correlation with the experimental data. The value of two parameters r_{max} and K_m appearing in Eq.(6) were calculated as 0.812 mg/

L.s and 0.1632 mg/L, respectively.

Using Eq.(7) one can find the variation of removal efficiency versus residence time. The plot of such a correlation was displayed in Fig.7. This figure shows that for an increase in removal efficiency from 95% to 99.999%, it is necessary to raise the retention time from 100 to 400 s. This fact demonstrates the capability of BTFR compared to wet scrubbers for high purification purposes. In wet scrubbers such an increase in removal efficiency requires an extreme rise in gas retention time, and hence the equipment size. Thus, BTFR is a more economical and effective equipment for formaldehyde removal when a highly-purifying system is desirable.

CONCLUSIONS

In this study formaldehyde was removed from air contaminated by both batch and continues systems. Both systems show a high ability to remove formaldehyde above 95%. The results demonstrates that the mass transfer in gas phase is a controlling step in overall formaldehyde removal, and in order to design a BTFR accurately, one needs a correlation for gas mass transfer coefficient with gas velocity. A developed model could show us that for high purification purposes BTFR is more effective and economical than wet scrubbers.

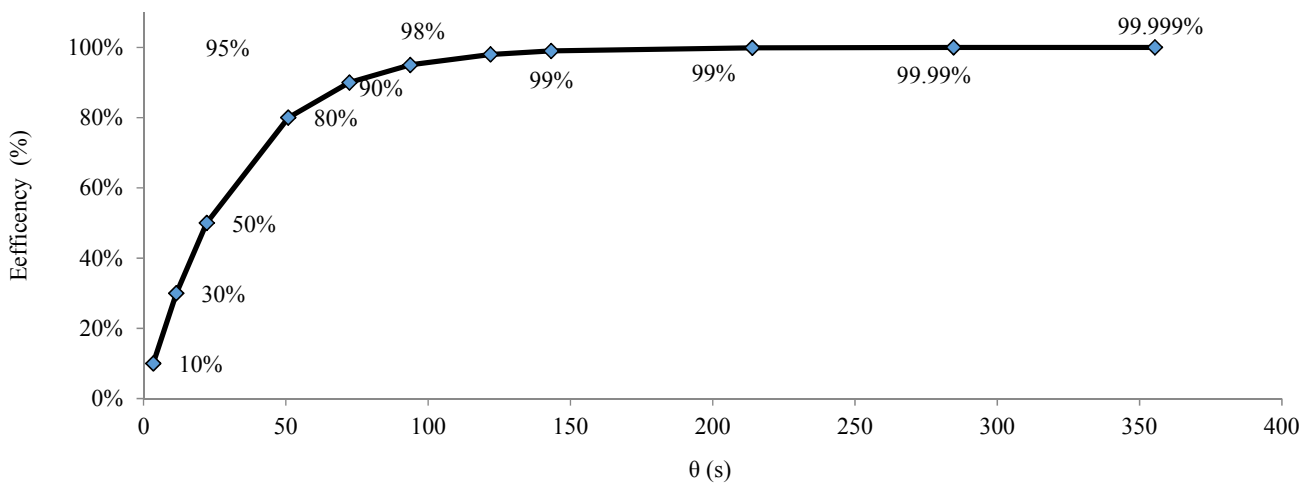


Fig.7. Model appropriate gas retention time of the BTFR system on a fictitious curve

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COMPETING INTERESTS

The authors declare that there is no conflict of interest that would prejudice the impartiality of this scientific work.

ETHICAL CONSIDERATIONS

Authors are aware of, and comply with, best practice in publication ethics specifically with regard to authorship (avoidance of guest authorship), dual submission, manipulation of figures, competing interests and compliance with policies on research ethics. Authors adhere to publication requirements that submitted work is original and has not been published elsewhere in any language.

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